

Development of Magnetism in Strongly Correlated Cerium Systems: Non-Kondo Mechanism for Moment Collapse

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We present an *ab initio* based method which gives clear insight into the interplay between the hybridization, the coulomb exchange, and the crystal-field interactions, as the degree of $4f$ localization is varied across a series of strongly correlated cerium systems. The results for the ordered magnetic moments, magnetic structure, and ordering temperatures are in excellent agreement with experiment, including the occurrence of a *moment collapse* of non-Kondo origin. In contrast, standard *ab initio* density functional calculations fail to predict, even qualitatively, the trend of the unusual magnetic properties.

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The difficulties and interest in treating strongly correlated electron systems, and the consequences of correlation effects on magnetic behavior in the transitional $4f$ or $5f$ localization regime, provide one of the central problems of condensed matter physics.¹⁻³ The transitional regime behavior is neither atomiclike nor itinerant. This gives rise to an extremely interesting range of phenomena, but also causes very great difficulties in treating the theory of these phenomena adequately, especially in a way providing the ability to predict the behavior of specific materials.¹⁻³ An adequate treatment requires treating the interelectronic coulomb interaction, i.e. the correlation effects, as constrained by exchange symmetry.⁴⁻⁶ In this letter, we demonstrate an approach for treating these difficulties in predicting the interesting and complex behavior of an important series of cerium compounds.

The isostructural (rock-salt structure) series of the cerium monpnictides CeX ($X = \text{P, As, Sb, Bi}$) and monochalcogenides ($X = \text{S, Se, Te}$) have become prototype model systems for study, because of their unusual magnetic properties.⁷⁻¹³ This series of strongly correlated electron systems offers the opportunity to vary systematically, through chemical pressure, the lattice constant and the cerium-cerium separation on going down the pnictogen or chalcogen column, and hence tailor the degree of $4f$ localization from the strongly correlated limit in the heavier systems to the weakly correlated limit in the lighter systems.⁷⁻¹³ The calculated single-impurity Kondo temperature, T_K , presented below, is much smaller than the magnetic ordering temperature in these systems, and hence this series lies in the *magnetic* regime of the Kondo phase diagram.¹⁴ Nevertheless, in this work we demonstrate that the sensitivity of the hybridization, coulomb exchange, and crystal-field interactions with the chemical environment gives rise to a variety of unusual and interesting magnetic properties across the series, in agreement with experiment, *including the occurrence of a non-Kondo magnetic moment collapse*.

This class of cerium systems exhibits large magnetic anisotropy which changes from the $\langle 001 \rangle$ direction in the pnictides to the $\langle 111 \rangle$ direction in the chalcogenides. The low-temperature ordered magnetic moment increases with increasing lattice constant for the pnictides from $0.80\mu_B$ in CeP to $2.1\mu_B$ in CeSb and CeBi,⁷⁻⁸ while it decreases with increasing lattice constant for the chalcogenides from $0.57\mu_B$ in CeS to $0.3\mu_B$ in CeTe.⁷⁻⁹ The *magnetic moment collapse* from CeSb to CeTe, with both systems having about the same lattice constant, is indicative of the sensitivity of the magnetic interactions to chemical environment. The experimentally observed low-temperature structure in CeBi and CeSb is the $\langle 001 \rangle$ antiferromagnetic type IA ($\uparrow\uparrow\downarrow\downarrow$), whereas in CeAs and CeP the structure is the $\langle 001 \rangle$ antiferromagnetic type I ($\uparrow\downarrow$).^{7,15} The ordering temperature increases from 8K in CeP to 26K in CeBi for the pnictides, whereas it decreases from 8.4K in CeS to an unusually low 2.2K in CeTe.⁷⁻¹¹ Another unusual feature of this series of cerium compounds is the large *suppression* of the crystal field (CF) splitting of the Ce^{3+} free-ion $4f_{5/2}$ multiplet from values expected from the behavior of the heavier isostructural rare-earth pnictides or chalcogenides.¹⁶ This can be understood¹⁷ as arising from band- f hybridization effects. In both the cerium monpnictides and monochalcogenides, the CF splitting between the Γ_7 doublet and the Γ_8 quartet decreases with increasing anion size, from 150 K for CeP to 10 K in CeBi and from 130 K for CeS to 30 K for CeTe, and it is about the same for the same row in both series, a rather surprising result in view of the additional valence electron on the chalcogen ion.¹⁸ Neutron scattering experiments have shown¹⁹ that the Γ_7 -doublet is the CF ground state in all the cerium pnictides and chalcogenides.

In this paper we present material-predictive results from two *ab initio* based methods to study the change of magnetic properties across this series of cerium systems. The first, *ab initio* based, method gives clear insight into the role of the three pertinent interactions: 1)

The band- f hybridization-induced inter-cerium magnetic coupling; 2) the corresponding effects of band- f coulomb exchange; and 3) the crystal-field interaction. This approach allows us also to understand the interplay between these interactions as the degree of $4f$ localization is varied across the series. The predictive calculations give results for the magnetic moments, magnetic structure, and ordering temperatures in excellent agreement with experiment. Thus, this approach allows to understand and predict a number of key features of observed behavior. First, is the very low moment and low ordering temperature of the antiferromagnetism observed in CeTe, an incipient heavy Fermion system. (For a review of theory and experimental behavior of heavy Fermion systems see references 2,3,20,21.) This *ab initio*-based method, described below, predicts the *magnetic moment and ordering temperature collapse* from CeSb to CeTe, both systems having about the same lattice constant but CeTe having an additional p electron. The origin of the moment collapse is of non-Kondo origin. The earlier work of Sheng and Cooper⁵ showed that this magnetic ordering reduction is accurately predicted without including any crystal-field effects. An erroneous statement appears in the recent review article by Santini *et al.*²² stating that crystal-field effects played an important role in the calculated results of Sheng and Cooper.⁵ This is incorrect, since crystal-field effects were *not* included in these calculations. We show in this paper that including the crystal-field effects modifies this behavior only quantitatively. Second, our results demonstrate that, while the band- f coulomb exchange mediated interatomic $4f$ - $4f$ interactions dominate the magnetic behavior for the heavier systems, which are more localized because of the larger Ce-Ce separation, the opposite is true for the lighter, more delocalized systems, where the hybridization-mediated coupling dominates the magnetic behavior. This reflects the great sensitivity of the relative importance of hybridization and coulomb exchange effects on magnetic ordering depending on the degree of $4f$ localization. Third, we show that for the lighter more delocalized systems the crystal-field interactions are much larger than the inter-cerium interactions and hence dominate the magnetic behavior. Finally, we predict the experimentally observed change of the ground-state magnetic structure from the $\langle 001 \rangle$ antiferromagnetic type IA ($\uparrow\uparrow\downarrow\downarrow$) in CeBi and CeSb to the $\langle 001 \rangle$ antiferromagnetic type I ($\uparrow\downarrow$) in CeAs and CeP. On the other hand, the second *ab initio* method, based on density functional theory within the local density approximation (LDA),^{23,24} fails to predict, even qualitatively, the trend of magnetic properties in this series of strongly correlated electron systems.

The first, *ab initio* based, method employs the degenerate Anderson lattice model which incorporates explicitly the hybridization and the coulomb exchange interactions on an equal footing^{4,5}

$$\begin{aligned}
H = & \sum_k \epsilon_k c_k^\dagger c_k + \sum_{Rm} \epsilon_m f_m^\dagger(R) f_m(R) \\
& + \frac{U}{2} \sum_{R, m \neq m'} n_m(R) n_{m'}(R) \\
& + \sum_{kmR} [V_{km} e^{-i\mathbf{k} \cdot \mathbf{R}} c_k^\dagger f_m(R) + H.C.] \\
& - \sum_{kk'} \sum_{mm'R} J_{mm'}(\mathbf{k}, \mathbf{k}') e^{-i(\mathbf{k}-\mathbf{k}') \cdot \mathbf{R}} c_k^\dagger f_m^\dagger(R) c_{k'} f_{m'}(R).
\end{aligned} \tag{1}$$

The parameters entering the model Hamiltonian, i.e., the band energies ϵ_k , the f -state energy ϵ_m , the on-site coulomb repulsion U , the hybridization matrix elements, V_{km} , and the band- f coulomb exchange $J_{mm'}(\mathbf{k}, \mathbf{k}') = \left\langle \phi_k^*(r_1) \psi_m^*(r_2) \left| \frac{1}{r_{12}} \right| \psi_{m'}(r_1) \phi_{k'}(r_2) \right\rangle$ are evaluated on a wholly *ab initio* basis from non-spin polarized full potential linear muffin tin orbital²³ (FPLMTO) calculations. Here, r_{12} stands for $|r_1 - r_2|$; ϕ_k are the non- f basis states of the FPLMTO, and ψ_m are the localized f states. Because the size of both the hybridization and coulomb exchange matrix elements are much smaller (~ 0.1 eV) than the intraatomic coulomb interaction U (6eV), one can apply perturbation theory and evaluate the anisotropic two-ion $6X6$ interaction matrices^{4,5}, $E_{m_1 m_1'}^{m_2 m_2'}(\mathbf{R}_2 - \mathbf{R}_1)$, which couple the two f -ions. The exchange interactions have three contributions: the wholly band- f coulomb exchange mediated interaction proportional to $J_{mm'}^2(\mathbf{k}, \mathbf{k}')$, the wholly hybridization-mediated exchange interaction proportional to V_{km}^4 , and the cross term proportional to $V_{km}^2 J_{mm'}(\mathbf{k}, \mathbf{k}')$. With the two-ion interactions having been determined, the low-temperature magnetic moment and the ordering temperature can be determined by use of a mean field calculation.^{4,5,8} We have previously applied this *ab initio* based method to investigate the effect of hybridization-induced cerium-cerium interactions^{4,17} and the combined effect of both the hybridization and coulomb induced interactions⁵ on the magnetic properties of the heavier cerium pnictides and chalcogenides (CeBi, CeSb, and CeTe). However, these calculations did not take into account the crystal field interaction and employed a warped muffin-tin LMTO calculation for the parameters entering the model. The excellent agreement found⁵ with experiment for the low-temperature magnetic moment and ordering temperature is relatively unaffected by the CF interaction, because the CF interaction in the heavier cerium systems is smaller than the two-ion exchange interactions.

The second method employs *ab initio* spin polarized electronic structure calculations based on the FPLMTO method²³ using 1) only spin polarization, with the orbital polarization included only through the spin-orbit coupling, and 2) both the spin and orbital polarization polarization.²⁴ In these calculations the $4f$ states are treated as band states. The orbital polarization is taken into account by means of an eigenvalue shift²⁴,

$\Delta V_m = -E^3 L_z m_l$, for the $4f$ atom. Here, L_z is the z -component of the cerium total orbital moment, m_l is the magnetic quantum number, and E^3 is the Racah parameter evaluated self-consistently at each iteration.

The crystalline field, which was neglected in the previous calculations,^{4,5} is expected to affect the magnetic behavior considerably, if it is large. It is important to emphasize that since in the first method the $4f$ states are treated as core states, they interact only with the spherical component of the effective one-electron potential. Thus, the interaction of the *atomic-like* $4f$ state with the *non-spherical* components of the potential, giving rise to the CF splitting, $\Delta_{CF} = \epsilon_{\Gamma_8} - \epsilon_{\Gamma_7}$, is not included in the calculation of the model Hamiltonian parameters. In this paper, we generalize the first, *ab initio* based, method to include both the interatomic $4f$ - $4f$ coupling and the crystal-field interactions on an equal footing and to employ a full potential LMTO evaluation of the model Hamiltonian parameters. While the effect of the full potential on both the hybridization and coulomb exchange interactions is small, including the CF interaction will be shown to play a role as important as the interatomic $4f$ - $4f$ interactions for understanding and predicting the *overall trend* in the unusual magnetic properties, as as one chemically tunes the degree of $4f$ localization across this series of strongly correlated electron systems. The resultant Hamiltonian is^{4,5}

$$H = - \sum_{i,j} \sum_{\substack{\mu,\nu \\ \epsilon,\sigma}} \xi_{\mu\nu}^{\epsilon\sigma}(\theta_{ij}) e^{-i(\mu-\nu+\epsilon-\sigma)\phi_{ij}} c_{\epsilon}^{\dagger}(j) c_{\sigma}(j) c_{\mu}^{\dagger}(i) c_{\nu}(i) \\ + B_4 \sum_i (O_4^0(i) + 5O_4^4(i)), \quad (2)$$

where the $\xi_{\mu\nu}^{\epsilon\sigma}(\theta_{ij})$ are the two-ion $4f$ - $4f$ interaction matrices rotated to a common crystal-lattice axis, and the O_4^0 and O_4^4 are the Stevens operators equivalents acting on the Ce^{3+} free-ion $4f_{5/2}$ multiplet.²⁵ The CF splitting is $\Delta_{CF} = 360B_4$; a positive B_4 value gives the Γ_7 ground state, which is experimentally observed.¹⁹ While our work in progress is aimed at evaluating the CF splitting on a wholly *ab initio* basis, in the absence of an *ab initio* value of the CF interaction in this class of strongly correlated cerium systems, the Δ_{CF} is set to the experimental values listed in Table 3.^{10,19}

In Table I, we present the calculated values of the zero-temperature cerium magnetic moment from the FPLMTO electronic structure calculations. Listed in the table are values both with and without the orbital polarization correction taken into account. Note, the importance of including the orbital polarization in these $4f$ correlated electron systems. As expected, in all cases, the orbital polarization is found to be opposite to the spin polarization. Comparison of the total energies predicts that the magnetic anisotropy changes from the $\langle 001 \rangle$ direction in the pnictides to the $\langle 111 \rangle$ in the chalcogenides, in agreement with experiment. On the other

TABLE I. Values of the calculated and experimental⁷⁻¹¹ low-temperature ordered magnetic moments for the cerium chalcogenides and pnictides in units of μ_B . Listed are the LMTO values for the spin moment μ_S , the orbital moment μ_L , and total moment μ , for the spin polarized only calculation and for the calculation with spin polarization and orbital polarization correction.

	FP+SP			FP+SP+OP			EXPT
	μ_S	μ_L	μ	μ_S	μ_L	μ	μ
CeS	-1.00	0.91	-0.09	-1.24	1.99	0.75	0.57
CeSe	-1.08	1.02	-0.06	-1.26	2.07	0.81	0.57
CeTe	-1.15	1.28	0.07	-1.31	2.29	0.98	0.30
CeP	-0.80	0.55	-0.25	-0.85	1.27	0.43	0.80
CeAs	-0.84	0.64	-0.20	-0.85	1.42	0.57	0.80
CeSb	-0.86	0.74	-0.12	-0.91	1.61	0.70	2.06
CeBi	-0.86	0.74	-0.12	-0.95	1.69	0.74	2.10

hand, except perhaps for the lighter chalcogenides (CeS and CeSe), comparison of the *ab initio* and experimental values for the magnetic moment indicates the *failure* of the LDA calculations to treat properly the correlation effects of the $4f$ states (treated as valence states) within the LDA as the degree of $4f$ correlations increases in the heavier pnictide systems. Furthermore, the *ab initio* calculations fail to predict the large *moment collapse* from CeSb to CeTe, the latter being described as an incipient heavy Fermion system.^{2,3,20,21}

In Table II, we list the values of the $m = m' = 1/2$ matrix elements (characteristic matrix elements of the $6X6$ exchange interaction matrix) for the first three nearest-neighbor shells for the light (CeP and CeS) and the heavier compounds (CeSb and CeTe). Listed separately in this table are the three contributions to the interatomic $4f$ - $4f$ interactions arising from band- f hybridization (V^4), band- f coulomb exchange (J^2), and the cross term. It is important to note that while the coulomb exchange mediated interactions dominate the magnetic behavior for the heavier, more localized, $4f$ systems, the opposite is true for the lighter, more delocalized, systems where the hybridization mediated interactions dominate the magnetic behavior. This change of behavior of the interatomic $4f$ - $4f$ interactions is a result of the sensitivity of the hybridization and coulomb exchange to the degree of $4f$ localization. Equally important, is that while both first and second nearest-neighbor $4f$ - $4f$ interactions are ferromagnetic for CeSb, there is an *interplay* between ferromagnetic first nearest-neighbor and antiferromagnetic second nearest-neighbor interactions for CeTe. (These interactions are mediated via scattering of conduction electrons). This results in a saturated ordered moment for CeSb and in the ordered *magnetic moment collapse* for CeTe (see Table III).

In order to determine whether the magnetic moment collapse might be of Kondo origin, we have evaluated the single-impurity Kondo temperature,²⁶ $k_B T_K$

TABLE II. Values of the $m = m' = 1/2$ matrix elements (characteristic matrix elements of the 6X6 interatomic $4f-4f$ interaction matrix $E_{m_a m'_a}^{m_b m'_b}(\mathbf{R}_b - \mathbf{R}_a)$, for the first, second, and third nearest-neighbor shells in degrees Kelvin. Listed are the values of the hybridization induced (E_{V^4}), cross terms (E_{V^2J}), and pure coulomb exchange (E_{J^2}) contributions.

	CeP			CeS		
	E_{V^4}	E_{V^2J}	E_{J^2}	E_{V^4}	E_{V^2J}	E_{J^2}
$R = (\frac{1}{2} \frac{1}{2} 0)$	2.23	0.64	1.53	0.85	-0.40	1.50
$R = (1 0 0)$	6.39	0.27	1.65	-1.60	0.04	-0.80
$R = (1 \frac{1}{2} \frac{1}{2})$	-0.08	-0.02	0.16	0.38	-0.16	0.13

	CeSb			CeTe		
	E_{V^4}	E_{V^2J}	E_{J^2}	E_{V^4}	E_{V^2J}	E_{J^2}
$R = (\frac{1}{2} \frac{1}{2} 0)$	0.70	0.34	7.30	0.17	-0.19	2.90
$R = (1 0 0)$	2.07	0.07	10.21	-0.19	0.04	-1.69
$R = (1 \frac{1}{2} \frac{1}{2})$	-0.02	-0.03	0.40	0.04	-0.06	-0.01

$= \text{De}^{-\frac{1}{2\rho(E_F)|J(E_F)|}}$, across the series. Here, D is the bandwidth of the conduction electron states, $\rho(E_F)$ is the density of states of the conduction electrons at the Fermi energy, and $J(E_F)$ is the conduction electron- f exchange interaction at the Fermi energy, which has contributions both from the coulomb exchange interaction in Eq. (2), provided that it is negative, and the hybridization-induced exchange interaction $|J_{hyb}(E_F)| = \frac{V^2(E_F)U}{(|E_f - E_F|)(|E_f - E_F| + U)}$, where $J_{hyb}(E_F) < 0$. We find that the coulomb exchange interaction in Eq. (2) evaluated at the Fermi energy is positive across the entire series and hence cannot give rise to the Kondo effect. Thus, only the hybridization-induced exchange interaction, $J_{hyb}(E_F)$, can give rise to the Kondo effect.²⁶ Using the *ab initio* values of the parameters entering the expression for T_K , we find that $T_K \ll T_{ord}$ across the entire series ($T_K < 10^{-4}\text{K}$). The Kondo temperatures in the monpnictide series is smaller than that in the chalcogenides, due to the fact that in the pnictides the Fermi energy lies in the pseudogap, resulting in low $\rho(E_F)$. These results suggest that the moment collapse from CeSb to CeTe is of non-Kondo origin. Rather, it results from an interplay of ferromagnetic and antiferromagnetic interatomic $4f-4f$ interactions which arises purely from differences in the underlying electronic structure.

Listed in Table III are the calculated zero-temperature ordered moment and ordering temperature, T_N , from the first, *ab initio* based, method, with and without the CF interaction. It is clear that for the heavier systems (CeBi, CeSb, CeTe) the effect of the CF interaction on the magnetic moments is small, and it is slightly more pronounced on the ordering temperatures. This is due to the fact that for the more localized systems the CF interaction is smaller than the two-ion interactions. This is the reason that the previous calculations,⁵ neglecting the CF interaction, gave results in very good agreement with

TABLE III. Calculated values (from the first *ab initio* based method) of the zero-temperature ordered moment μ in μ_B , and the ordering temperature T_N in degrees Kelvin, with and without the crystalline field (CF) interaction across the cerium pnictide and chalcogenides series. Also listed are the experimental values⁷⁻¹¹ of μ and T_N , and the CF splitting¹⁹ between the Γ_7 ground state and the Γ_8 state in degrees Kelvin.

	Δ_{CF}	μ_0			T_N		
		no CF	CF	exp	no CF	CF	exp
CeS	140	1.80	0.73	0.57	1.0	11.0	8.4
CeSe	116	1.10	0.79	0.57	2.5	14.0	5.7
CeTe	32	0.60	0.46	0.30	8.0	5.0	2.2
CeP	150	2.10	0.73	0.81	14	11	8
CeAs	137	2.10	0.74	0.85	16	13	8
CeSb	37	2.10	1.80	2.06	20	18	17
CeBi	8	2.10	2.10	2.10	40	40	26

experiment. On the other hand, for the lighter more delocalized systems the CF interactions are much larger than the interatomic $4f-4f$ interactions, and hence dominate the magnetic behavior. The overall decrease of the magnetic moments in the presence of the CF interaction in all systems, arises from the mixing of the off-diagonal angular momentum states $|\pm 5/2\rangle$ and $|\mp 3/2\rangle$ states from the CF interaction with Γ_7 ground state. Overall, we find that the first, *ab initio* based, approach which takes into account all three pertinent interactions (hybridization, coulomb exchange, and CF interactions) on an equal footing, yields results for both the zero-temperature moment and the ordered temperature (a more stringent test for the theory) in excellent agreement with experiment.

A final corroboration of the success of the first *ab initio* based method is that it predicts the experimentally observed change of the ground-state magnetic structure from the $\langle 001 \rangle$ antiferromagnetic type IA ($\uparrow\uparrow\downarrow\downarrow$) in CeBi and CeSb to the $\langle 001 \rangle$ antiferromagnetic type I ($\uparrow\downarrow$) in CeAs and CeP.^{7,15} More specifically, the sign [ferromagnetic (F) or antiferromagnetic (AF)] of the $|\pm 5/2\rangle$ matrix elements of the 6X6 exchange matrix determines the *interplanar* interaction between successive (001) Ce planes. We find, that for the heavier compounds (CeBi and CeSb) the $|\pm 5/2\rangle$ matrix elements of the *coulomb exchange* matrix are FM and hence favor the $\uparrow\uparrow\downarrow\downarrow$ type, while in the lighter systems (CeAs and CeP) the $|\pm 5/2\rangle$ matrix elements of the *hybridization*-induced two-ion matrix are AF, and hence they favor the $\uparrow\downarrow$ type.

In conclusion, we have applied two different, *ab initio* based and *ab initio* LDA, methods to study the dramatic change of magnetic properties across a series of strongly correlated electron systems which offer the opportunity to chemically tailor the different type of interactions (band- f hybridization, band- f coulomb exchange, and CF interactions), pertinent to the unusual magnetic

behavior. The first, *ab initio* based, approach which explicitly takes into account the interplay of the three pertinent interactions, gives results in excellent agreement with experiment for all compounds in the series, including the *moment collapse* from CeSb to CeTe and the trend of moments and ordering temperatures across the series. The remaining problem of determining on a wholly *ab initio* basis the *suppressed* crystal-field interactions in this class of systems poses a theoretical challenge for future theoretical work. On the other hand, the second, fully *ab initio* LDA, method gives good results for the lighter chalcogenide systems, but it entirely fails to give, even qualitatively, the trend of the unusual magnetic behavior.

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